We thank reviewer #1 for taking the time evaluating our manuscript. We provided a point-by-point response to the reviewer's comments given in bold faced letters below.

The water activity is a key variable for parameterization of the results; aw is a function of the concentration, temperature and pressure. In equilibrium with water vapor aw is equivalent to the ambient relative humidity (RH). The authors need to explain in more detail how they control the RH during the experiments. They mention that RH is derived from the dew point temperature and drop temperature; however, the temperature of the system varies during the runs (10K/min). The uncertainty of aw needs to be addressed in detail as well.

We intended to keep the experimental description short since it has been published multiple times (e.g. Knopf and Lopez, 2009, Knopf et al., 2011, Alpert et al., 2011a,b, Knopf and Rigg, 2011). In short, RH and thus a_w is set at the beginning of the experiment as the sample is introduced into the aerosol conditioning cell (ACC). The droplets are conditioned by exposure to constant relative humidity, defined by water partial pressure given by the dew point temperature and the temperature of the droplets. This sets droplet a_w with given uncertainty. Then the droplets are sealed off against ambient air while remaining in the ACC. After being sealed they are transferred to the ice nucleation setup. During the ice nucleation experiment (as the temperature cools with 10 K min^{-1}) the droplets are not exposed to humidified gas. The aerosol cell is designed in such a way that the amount of potentially condensing water vapour within this cell upon cooling is negligible compared to the condensed phase water within the droplets following previous experimental studies (see studies above and Koop et al., 1998, Knopf et al., 2002). Thus droplet composition as set in the ACC stays constant throughout the ice nucleation experiment. Water activity does not change significantly with decreasing temperature in the case of applied aqueous ammonium sulphate solutions as shown in previous literature (Bertram et al., 2000, Knopf and Lopez, 2009 and others) and by thermodynamic models such as E-AIM (Clegg et al., 1998). Therefore the uncertainty in a_w stays the same as derived from the uncertainty from the conditioning procedure.

We add the following sentences to the text for clarification:

p. 4923, l. 13: "Droplet conditioning is conducted at droplet temperatures of 290-295 K."

p. 4923, l. 20: "Hence, droplet composition is fixed for the remainder of the experiments according to the conditioning procedure. Subsequent ice nucleation experiments in which the aerosol sample is cooled, will not significantly change droplet composition since the amount of water vapour within the sealed aerosol sample that can condense onto the droplets is negligible compared to the amount of the condensed phase water (Koop et al., 1998, Knopf et al. 2002, Knopf and Lopez, 2009)."

p. 4924, l. 24: "For aqueous $(NH_4)_2SO_4$ solutions it has been shown experimentally and by thermodynamic models (Bertram et al., 2000, Knopf and Lopez, 2009, Clegg et al., 1998) that a_w does not change significantly with temperature for investigated a_w range. For this reason, droplet a_w and corresponding uncertainty as defined by the droplet conditioning procedure apply to the ice nucleation experiments observed at lower temperatures."

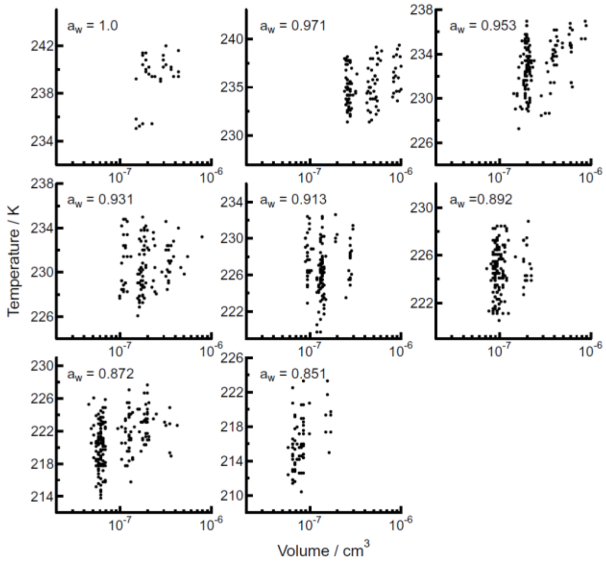
It is important to show the droplet size distribution used in the experiments. Is the droplet spectrum representative of droplets found in the upper region of the atmosphere where the nucleation processes occur? A figure with the droplet size distribution should be included in the paper and the influence of the droplet size in the freezing temperature should be discussed.

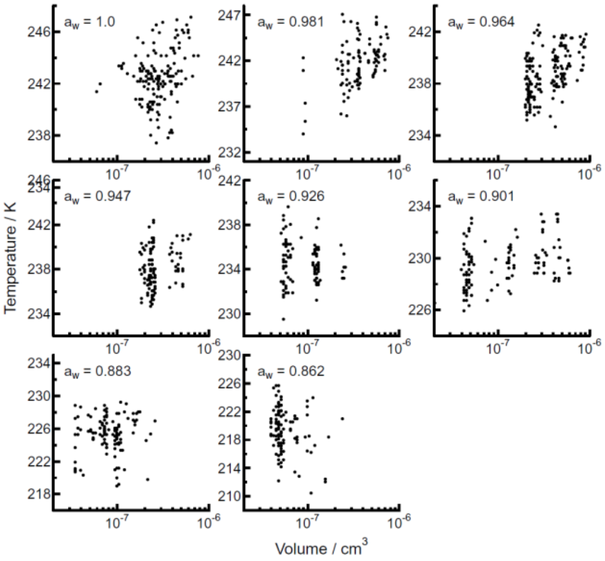
Applied droplets are larger in size than the ones we would expect to nucleate ice via immersion freezing in the upper atmosphere. We will add this information in the introduction.

p. 4921, l. 15: We change the original sentence "In this study we investigate immersion freezing with respect to temperature and nucleation kinetics from micrometer-sized water and aqueous $(NH_4)_2SO_4$ droplets containing PP and LEO particles acting as IN for a_w of 0.85–1.0 and temperatures from 273–215 K."

to "In this study we investigate immersion freezing with respect to temperature and nucleation kinetics from micrometer-sized water and aqueous $(NH_4)_2SO_4$ droplets, representing slightly larger droplets than observed in the atmosphere (Sassen and Dodd, 1988, Heymsfield et al., 1998, Ansmann et al., 2008), containing PP and LEO particles acting as IN for a_w of 0.85–1.0 and temperatures from 273–215 K."

Regarding the droplet size spectrum and its effect on freezing, we apply in principle monodisperse droplets as discussed in the sample preparation. The piezo-electric droplet dispenser should provide monodisperse droplets on a single droplet basis, however, there can be slight variations in droplet diameters. Depending on applied relative humidity, the droplets take up more or less water resulting in additional differences in droplet diameters even so the initial droplet size generated by the dispenser is the same. As can be seen from new figures S2 and S3 droplet volumes are reasonably within one order of magnitude. Furthermore, the figures indicate no freezing temperature dependence on droplet volume as inferred in our previous studies (Alpert et al., 2011a).





New Figs. S2 and S3 presenting heterogeneous freezing temperatures of water and aqueous $(NH_4)_2SO_4$ droplets containing Leonardite and PP particles shown as dots are plotted as a function of droplet volume for each investigated water activity.

We would not expect that the droplet volume affects the freezing process. Water molecules and their respective hydrogen bonding network "move/fluctuate" on time scales of sub-picoseconds (McGuire and Shen, 2006). In these time scales water does not move long distances. Thus water molecules at the surface of the IN forming critical clusters do not "know" of the actions of water molecules nm or µm away. Also, a larger aqueous volume will not increase the likelihood of homogeneous ice nucleation. For example, at median freezing temperatures of Pahokee peat J_{het} = 5E4 cm^{-2} s^{-1}. At these temperatures J_{hom} = 1.55E-10 cm^{-3} s^{-1} (Koop et al. 2000). Homogeneous ice nucleation becomes competitive only if the surrounding volume would be > 1E8 cm^3 of aqueous solution – clearly not atmospherically relevant.

We add the requested figures to the supplement (new Figs. S2 and S3) and add the following text to the manuscript, p. 4925, l. 19:

"Furthermore, we analyzed immersion freezing temperatures as a function of aqueous droplet volume. As shown in Figs. S2 and S3, no dependence of freezing temperatures on droplet volumes for LEO and PP is observed. This is similar to our previous findings by Alpert et al. (2011a). This finding may not be surprising when considering that water at interfaces fluctuates on time scales of sub-picoseconds (McGuire and Shen, 2006), implying that the water clusters at the IN surface are not influenced by water further away within the droplet. Also, the homogeneous ice nucleation rate is negligible at these heterogeneous freezing temperatures and droplet volumes (Koop et al., 2000)."

The authors say "*However, for PP, the SEM based surface area estimates are about a factor of 8 larger than the ones derived from BET analysis. The reason for this difference may be due to the partial solubility of PP. For the remainder of this work we apply the BET obtained surface areas for ice nucleation analysis. The reason for this difference may be due to the partial solubility of PP. For the remainder of this work we apply the BET obtained surface areas for ice nucleation analysis.*" The reason for the difference is not clearly stated. The reasons for choosing the surface areas obtained with BET instead SEM need to be stated. Data analysis considering both results (BET and SEM) should be performed in order to know the variability of the results in terms of the surface areas.

As given in table 2 we have stated the uncertainties in both applied methods. For BET this represents the instrumental uncertainty and for the visual inspection and estimation, this is derived from statistical analysis of several investigated droplet samples. Also, in response to referee #3, the difference in surface area between BET and SEM measurements are attributed to the nature of the technique used to obtain the results. For BET, the PP and LEO remain anhydrous and thus their macromolecular size and orientation remains mostly unchanged. For SEM these particles are placed in water, and as a result parts of these macromolecules dissolve or rearrange thereby altering the size and orientation with subsequent effects on the visible condensed phase surface area. Also, in response to reviewer #2, sonication my lead to a breakup of particulate aggregates. The BET measurements are, as stated, in line with previous BET measurements of humic acids.

We change the sentence on p. 4924, l. 11:"The reason for this difference may be due to the partial solubility of PP." to
"A possible reason for this difference is that PP and LEO remain anhydrous when examined by BET and thus their macromolecular size and orientation remains mostly unchanged. However, for SEM derived surface areas, these particles are placed in water and parts of these macromolecules may dissolve or rearrange thereby altering their size, orientation, or lead to breakup of particulate aggregates with subsequent effects on the visible condensed phase surface area."

A cooling rate of 10 K/min was used in this work. Several works in literature point out the relevance of this parameter in the nucleation processes. In order to check the influence of this parameter in the current study, the authors should extend their measurements to different cooling rates. The same comment is valid for heating rate.

We agree that investigating the cooling rate dependence to infer the time dependency of the ice nucleation process would be beneficial. However, in this study our main focus was on investigating immersion freezing as a function of water activity and to infer if previously applied ice nucleation parameterisations can capture the effect of water activity – not previously done. In a subsequent manuscript, which was just accepted (Knopf and Alpert, 2013), we have addressed these points by determining the immersion freezing points for selected water activities and cooling rates of 1, 5, 9.5, 14.2 K min^{-1}. We observe a dependency of the freezing points with varying cooling rates as expected from classical nucleation theory. Discussion of these findings would be beyond the scope of this work.

The heating rate is chosen in such a way to give us the most accurate melting point determination under reasonable experimental time scales. Accurate melting points are necessary to validate water activity/composition conditioning of the droplets and to rule out composition changes during the ice nucleation experiments. If the ice melting temperature does not agree with the ice melting curve as a function of water activity (Koop et al., 2000), then the water activity of the droplets is not correct and the sample is discarded. The melting rate is deduced from our calibration experiments by measuring the melting point of similar sized ice crystals and melting points of various organic solutes. Clearly, fast heating rates (e.g. 10 K min-1) will result in overestimated melting temperatures since the temperature within the droplet may not follow the substrate temperature. A rate of 0.5 K min^{-1} represents a very good compromise within experimental temperature uncertainty and time scales.

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